

# Phonons in Random Elastic Media and the Boson Peak

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We show that the density of states of random wave equations, normalized by the square of the frequency, has a peak — sometimes narrow and sometimes broad — in the range of wave vectors between the disorder correlation length and the interatomic spacing. The results of this letter may be relevant for understanding vibrational spectra and light propagation in disordered solids.

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One of the intriguing features of random elastic media — observable in both Raman or neutron cross sections but also in calorimetric measurements — is an anomalous accumulation of phonons at low frequencies [1]. This phenomenon finds its most prominent manifestation in a peak in the quantity  $I(\omega) = \rho(\omega)/\omega^2$ , where  $\rho(\omega)$  is the phonon density of states (DoS) and  $\omega$  is the frequency. In recent years, many competing theories as to the origin of this so-called boson peak have been formulated (for a recent reference, see Ref. [2] and references therein). Of these approaches a majority is based on model mechanisms specific to the low temperature physics of amorphous solids. Yet the boson peak is shown by both glassy and random crystalline systems alike, an observation which has ignited the search for an explanation which is not tied to the specifics of a glassy environment.

On a basic level, the acoustic excitations of both amorphous materials and disordered crystals are described by random wave equations. Existing analyses of such equations in the literature indeed predicted a disorder generated excess DoS, [3]. However, these structures were observed at high frequencies (wavelengths of the order of the interatomic spacing), while the boson peak is a low energy phenomenon. In this letter we argue that the DoS of elastic vibrations in disordered media is enhanced by disorder at low frequencies, with  $I(\omega)$  exhibiting either a peak or a broad maximum (see Fig. 1 for a representative picture of the DoS). Which of the two will be observed crucially depends on the modelling of the randomness, i. e. the *type* of disorder at work. Since the profile of the randomness of ‘real’ systems is generally unknown, we are not in a position to judge whether the present mechanism alone may account for the spectral peaks observed in all experiments. We believe, however, that it operates under quite general conditions.

Let us begin by discussing some generic large scale structures of the DoS of acoustic excitations. In a clean system, the linear relation  $\omega = \bar{c}k$  between frequency and

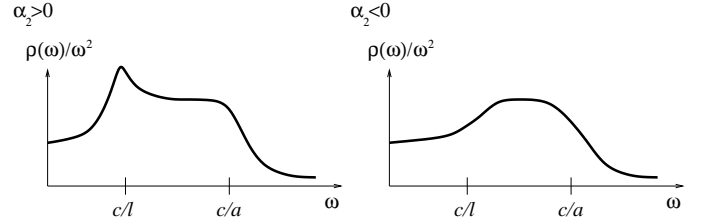


FIG. 1:  $I(\omega)$  for the random elastic media in cases where  $\alpha_2 > 0$  or  $\alpha_2 < 0$ .

magnitude  $k$  of the wave vector gives rise to a DoS

$$\rho(\omega) = \int \frac{d^d k}{(2\pi)^d} \delta(\omega - \bar{c}k) = \frac{A_d}{(2\pi\bar{c})^d} \omega^{d-1}. \quad (1)$$

Here,  $A_d$  is the area of the  $d$ -dimensional unit sphere and  $\bar{c}$  the speed of sound (for our present analysis, the potential existence of several acoustic phonon branches with different sound velocities will not be of importance). Tied to the linearizability of the dispersion relation, the relation  $\rho \sim \omega^{d-1}$  applies only to frequencies  $\omega \ll \omega_D$  much smaller than the Debye frequency  $\omega_D \sim \frac{\bar{c}}{a}$  ( $a$ : lattice spacing.) On dimensional grounds, the generalization of (1) to higher frequencies must be of the form

$$\rho(\omega) \sim \omega^{d-1} \tilde{f}(\omega/\omega_D), \quad (2)$$

where  $\tilde{f}(u)$  is some function with  $\tilde{f}(u \ll 1) \simeq 1$ . It is clear that  $\tilde{f}(u)$  falls off to zero at  $u \gg 1$ .

In contrast, in disordered materials we find strong deviations from the above scaling form already for frequencies  $\omega \ll \omega_D$ . Indeed, our analysis of the random wave equation below will lead us to

$$\rho(\omega) = \frac{A_d}{(2\pi\bar{c})^d} \omega^{d-1} f\left(\frac{\omega l}{\bar{c}}\right), \quad \omega \ll \omega_D, \quad (3)$$

where the effective correlation length  $l \gg a$  of the disorder is assumed to be much larger than the lattice spacing, and  $\bar{c}$  is the typical speed of sound in random media to be defined more precisely below. The scaling function  $f$

is given by

$$\begin{aligned} f(u) &= 1 + \alpha_1 u^2 + \dots, \quad u \ll 1, \\ f(u) &= \beta \left( 1 + \frac{\alpha_2}{u^2} + \dots \right), \quad 1 \ll u \ll \frac{l}{a}, \end{aligned} \quad (4)$$

where the form of the coefficients  $\alpha_{1,2}$ ,  $\beta$  depends on the space dimensionality and on a few basic characteristics of the randomness. In the most interesting case,  $d = 3$ , we find that  $\beta > 1$  and  $\alpha_1 > 0$ . The sign of  $\alpha_2$  lacks universality.  $\alpha_2$  is negative if disorder in materials is mostly in the random elastic constants, and it is positive if disorder is mostly due to fluctuating mass density.

For  $\alpha_2 > 0$ ,  $I(\omega)$  grows for  $\omega \ll \bar{c}/l$  and falls off at  $\omega \gg \bar{c}/l$  (while still  $\omega \ll \omega_D$ ). This implies the existence a peak at  $\omega \sim \bar{c}/l$ . In contrast, for  $\alpha_2 < 0$ ,  $I(\omega)$  increases until  $\omega$  becomes much larger than  $\bar{c}/l$ . Combined with the expected drop-off of  $I(\omega)$  at  $\omega \gtrsim \omega_D$ , this produces a broad maximum for  $I(\omega)$  between  $\bar{c}/l$  and  $\omega_D$ . Fig. 1 shows a caricature of the two scenarios. We finally note that if the disorder is relatively weak,  $\text{rms } c < \bar{c}$ , (so that the fluctuations of local speed of sound  $c$  are smaller than the typical sound velocity)  $\alpha_1 \sim \alpha_2 \sim \text{var } c/\bar{c}^2$ ; for more generally applicable expressions, see below.

We next turn to the derivation of these results. Consider the wave equation

$$\left( \Delta + \frac{\omega^2}{c^2(\mathbf{x})} \right) \psi(\mathbf{x}) = 0, \quad (5)$$

where the random velocity field  $c(\mathbf{x})$  fluctuates on spatial scales  $\sim l$ . Interpreting the variable  $m(\mathbf{x}) \equiv 1/c^2(\mathbf{x})$  as the mass density of the random medium, we refer to Eq. (5) as a ‘random mass’ wave equation.

In the regime of small frequencies,  $\omega \ll \bar{c}/l$ , we are facing a situation where the correlation length  $l$  is much smaller than the ‘typical’ wave length  $k^{-1} \equiv \bar{c}/\omega$  at which the wave function  $\psi(\mathbf{x})$  fluctuates. The phonon field effectively averages over many fluctuation intervals of the disorder and, to a first approximation, Eq. (5) may be replaced for its average over random  $c(\mathbf{x})$ :  $\left( \Delta + \frac{\omega^2}{\bar{c}^2} \right) \psi(\mathbf{x}) = 0$ , where the typical speed of sound  $\bar{c}$  is defined as

$$\bar{c} \equiv \langle c(\mathbf{x})^{-2} \rangle^{-1/2} \quad (6)$$

and the angular brackets denote averaging over random  $c(\mathbf{x})$ . The very low frequency asymptotics is then given by Eq. (1) with  $\bar{c}$  taken from Eq. (6). (This result, as well as the high frequency asymptotics Eq. (10) discussed below, are originally due to Chalker [4].)

To compute corrections to the low frequency asymptotics of the DoS we rewrite Eq. (5) as

$$\left( -\Delta - \frac{\omega^2}{\bar{c}^2} h(\mathbf{x}) \right) \psi(\mathbf{x}) = \frac{\omega^2}{\bar{c}^2} \psi(\mathbf{x}), \quad (7)$$

where the function  $h(\mathbf{x}) \equiv [\bar{c}/c(\mathbf{x})]^2 - 1$  describes the randomness. This representation suggests to interpret  $\omega^2/\bar{c}^2$

as the eigenvalue of the operator  $\Delta$  weakly perturbed by  $\omega^2 h(\mathbf{x})/\bar{c}^2$ . The unperturbed problem is trivially diagonalized by a set of plane waves  $\psi_{\mathbf{k}}(\mathbf{x}) \equiv \frac{1}{L^{d/2}} \exp(i\mathbf{k} \cdot \mathbf{x})$ , with eigenvalues  $\omega^2/\bar{c}^2 = k^2$  ( $L$  is the system size.) We next apply standard perturbation theory to compute the eigenvalue shift caused by the presence of the perturbation  $\omega^2 h/\bar{c}^2$ . The vanishing of  $\langle h \rangle$  implies that, on average, there are no first order corrections. To second order in  $h$  we find that the average eigenvalue is given by  $\frac{\omega^2}{\bar{c}^2} = k^2 + k^4 \int \frac{d^d k'}{(2\pi)^d} \frac{g(\mathbf{k}')}{k'^2}$ , where  $g(\mathbf{k})$  is the Fourier transform of the disorder correlation function of  $g(\mathbf{x} - \mathbf{y}) \equiv \langle h(\mathbf{x})h(\mathbf{y}) \rangle$ . Irrespective of the distribution of the disorder, this function (a) drops off rapidly for  $k > l^{-1}$  and (b) approaches a constant value  $\sim l^d \text{var}(h)$  for  $k \ll l^{-1}$ . This implies that for  $d > 2$ , the integral above is dominated by momenta  $k' \sim l^{-1}$ . At the same time, the reference momentum  $k \sim \omega/\bar{c} \ll l^{-1}$  is small. We thus neglect the  $k$ -dependence of the integrand and arrive at

$$\frac{\omega^2}{\bar{c}^2} = k^2 - k^4 \int \frac{d^d k'}{(2\pi)^d} \frac{g(\mathbf{k}')}{k'^2}. \quad (8)$$

Physically, the correction to the zeroth order eigenvalue is due to virtual scattering events wherein states with low-lying momentum  $k \sim \omega/\bar{c}$  scatter off rapid fluctuations of  $h$  into high-lying states  $k' \sim l^{-1}$ . Yet for small frequencies, the large phase volume  $\sim l^{-d}$  available to these scattering processes cannot outweigh the overall multiplicative factor  $\omega^4$ . This mechanism pervades to higher orders in the eigenvalue expansion and justifies the perturbative approach. In particular, Eq. (8) indeed describes the dominant correction to the low frequency dispersion relation.

We next substitute Eq. (8) into  $\rho(\omega) = \frac{1}{L^d} \sum_{\mathbf{k}} \delta(\omega(k) - \omega)$  to obtain an expansion of the DoS as in Eqs. (3,4). Specifically, the coefficient

$$\alpha_1 = \frac{d+2}{2l^2} \int \frac{d^d k}{(2\pi)^d} \frac{g(\mathbf{k})}{k^2} \sim \text{var}(h) > 0, \quad (9)$$

where the proportionality to  $\text{var}(h)$  follows from the properties of the correlation function  $g$  discussed above. Interestingly, positivity of  $\alpha_1$  in Eq. (9) is a direct consequence of the fact that the second order perturbation theory always lowers the ground state energy.

For small velocity fluctuations,  $\text{var}(c) \ll \bar{c}^2$ , we obtain the estimate  $\alpha_1 \sim \text{var}(h) \sim \text{var}(c)/\bar{c}^2$  quoted above. We finally note that these results hold only for  $d > 2$ . For  $d \leq 2$ , the integral in Eq. (9) is infrared divergent and the approximation scheme employed here breaks down.

We next turn to the discussion of the high frequency case,  $\omega \gg \bar{c}/l$ . In this regime, the velocity field varies very little over length scales comparable to the typical wavelength. This implies that, locally, the solutions of (5) behave like plane waves with the local dispersion relation

$k(\mathbf{x}) = \omega/c(\mathbf{x})$  and the local density of states given by

$$\rho(\omega, \mathbf{x}) = \frac{A_d}{(2\pi)^d c^d(\mathbf{x})} \omega^{d-1}.$$

Averaging this result over the random velocity field we obtain the global density of states

$$\rho(\omega) = \frac{A_d}{(2\pi)^d} \left\langle \frac{1}{c^d(\mathbf{x})} \right\rangle \omega^{d-1}, \quad \omega \gg \frac{\bar{c}}{l}.$$

We thus find that the coefficient  $\beta$ , introduced in Eq. (4), is given by

$$\beta = \frac{\langle c^{-d}(\mathbf{x}) \rangle}{\langle c^{-2}(\mathbf{x}) \rangle^{\frac{d}{2}}}. \quad (10)$$

As a consequence of the convexity of the power function,  $\beta > 1$  for  $d > 2$ .

To obtain corrections to the high frequency asymptotics (10), we need to compute distortions in the spectral density caused by shallow (compared to the wave length) variations of the velocity field. It turns out that this task is most efficiently tackled by analyzing the Green's function

$$G = - \left( \frac{1}{\omega^2} \nabla^2 + m(\mathbf{x}) + i\epsilon \right)^{-1}, \quad (11)$$

where  $m \equiv c^{-2}$ . From (11) the average DoS is obtained as

$$\rho(\omega) = \frac{2}{\pi\omega} \text{Im} \langle m(\mathbf{x}) G(\mathbf{x}, \mathbf{x}) \rangle, \quad (12)$$

What makes the operator (11) a good starting point for our analysis is its structural similarity to a Schrödinger operator with ‘Planck’s constant’  $\hbar \sim \omega^{-1}$ . This analogy will enable us to apply semiclassical approximation schemes familiar from quantum mechanics. We begin by applying the Wigner transform

$$G(\mathbf{x}, \mathbf{y}) = \frac{\omega^d}{(2\pi)^d} \int d^d k G(\mathbf{x}, \mathbf{k}) e^{i\omega \mathbf{k} \cdot (\mathbf{x} - \mathbf{y})},$$

whereupon the ‘Schrödinger equation’ assumes the form

$$\left[ (\mathbf{k} - i\omega^{-1} \partial_{\mathbf{x}})^2 - m(\mathbf{x}) - i\epsilon \right] G(\mathbf{x}, \mathbf{k}) = 1. \quad (13)$$

To make use of the smallness,  $\sim \omega^{-1}$ , of the derivative operators, we expand  $G(\mathbf{x}, \mathbf{k})$  in powers of  $\omega^{-1}$ ,

$$G(\mathbf{x}, \mathbf{k}) = G^{(0)}(\mathbf{x}, \mathbf{k}) + \frac{1}{\omega} G^{(1)}(\mathbf{x}, \mathbf{k}) + \frac{1}{\omega^2} G^{(2)}(\mathbf{x}, \mathbf{k}) + \dots,$$

substitute the expansion into Eq. (13), and find  $G^{(0)}$ ,  $G^{(1)}$ ,  $G^{(2)}$ , ..., recursively. By symmetry,  $G^{(1)}$  vanishes upon configurational averaging so that the dominant correction to the DoS is provided by  $G^{(2)}$ . Substituting this

coefficient into Eq. (12), and comparing with Eq. (4) we obtain the result

$$\alpha_2 = \frac{(d-2)^2 (4-d)}{24} \frac{l^2}{\bar{c}^2 \langle c^{-d} \rangle} \left\langle \frac{\nabla c \cdot \nabla c}{c^d} \right\rangle.$$

Clearly,  $\alpha_2 > 0$  if  $d = 3$ . If the fluctuations of  $c$  are weak,  $\alpha_2 \sim \text{var } c / \bar{c}^2 \sim \alpha_1$ .

We thus derived Eq. (4). The positivity of the two expansion coefficients  $\alpha_{1,2}$  implies that the scaling function  $f(u)$  in Eq. (3) grows at  $u \ll 1$  and falls off at  $u \gg 1$ . This implies that the functional profile of the DoS contains a peak somewhere at  $u = \mathcal{O}(1)$ .

To conclude our analysis of the prototypical wave equation (5), let us briefly discuss the case of space dimensions  $d \neq 3$ . For  $d = 1$ , the high frequency coefficient  $\alpha_2 > 0$ , yet  $\beta < 1$ . Due to the instability in the low frequency expansion, the methods applied in this letter do not allow to calculate  $\alpha_1$  at  $d \leq 2$ . However, for certain distributions of the disorder transfer matrix methods may be applied to obtain an exact solution [5]. These calculations show that  $f(u)$  is a globally decreasing function, and  $\alpha_1 < 0$ . For  $d = 2$ , the two dominant high-frequency coefficients are structureless,  $\beta = 1$  and  $\alpha_2 = 0$ , while the low frequency expansion in Eq. (4) will now involve terms proportional to  $\log(u)$ . From these results we cannot decide whether  $\rho(\omega)/\omega$  has a maximum or a minimum between its low and high frequency asymptotics. Finally, for  $d > 3$ ,  $\alpha_1 > 0$  and  $\beta > 1$ , however  $\alpha_2 \leq 0$ , which implies that  $f(u)$  is a monotonously increasing function. Summarizing, we see that the normalized DoS of Eq. (5),  $\rho(\omega)/\omega^{d-1}$ , exhibits a peak at wavelengths of the order of disorder correlation length only for  $d = 3$ .

Before proceeding, let us briefly compare our so far results to earlier work. In most numerical simulations of the problem (cf. e.g., Ref. [3] and references therein), the disorder is chosen to be uncorrelated, that is, its correlation length is of the order of the lattice spacing. For such type of disorder,  $u \simeq 1$  translates to  $\omega \sim \omega_D$  deep in the bulk of the spectrum. At these frequencies it is hard to tell whether deviations from the low frequency asymptotics  $\rho(\omega) \sim \omega^2$  are caused by lattice effects (cf. Eq. (2)) or by disorder (Eq. (3)). Qualitatively, however, the numerical data is in agreement with the results of our present analysis. Turning to earlier analytical work, we notice that most approaches to random elastic problems rely on the self consistent Born approximation (SCBA). (See Ref. [6] for a general review of the methods involved and Refs. [7, 8] for the applications of these methods to random wave equations.) However, for a number of reasons the results obtained by this method lack quantitative reliability when used to calculate the DoS: (i) Within the standard SCBA,  $1/c^2$  is modelled as a Gaussian distributed variable. This implies the existence of rare domains where  $1/c^2$  is negative. Using the techniques introduced in Ref. [9] it is possible to show that these domains, no matter how small, lead to unstable

wave modes and to unphysical sharply growing contributions to the density of states at higher frequencies. (ii) The SCBA neglects certain contributions ('crossed diagrams') to the perturbative expansion of the Green function (11). At high frequencies, this approximation becomes invalid. The value of the threshold frequency beyond which the SCBA breaks down depends on the type of disorder under consideration. At any rate, however, it is parametrically smaller than the band width. At the same time, (iii), previous studies focused on the case of short range correlated disorder. In this case, deviations of the scaling law  $\rho \sim \omega^{d-1}$  are expected at frequencies  $\omega \sim \omega_D$ , i.e. in a regime where the SCBA breaks down and lattice effects intervene. (The alternative techniques in this letter avoid all of these problems.)

The wave equation Eq. (5) applies to the specific case, where only the mass density of the elastic medium fluctuates. The continuum description of a more general environment wherein the elastic constants also fluctuate reads as

$$(\nabla \mu(\mathbf{x}) \nabla + \omega^2 m(\mathbf{x})) \psi(\mathbf{x}) = 0. \quad (14)$$

Here both  $\mu(\mathbf{x})$  and  $m(\mathbf{x})$  are random positive quantities.

It turns out that the direct perturbative expansion applied above to Eq. (5) cannot be used to determine the coefficient  $\alpha_1$  of the problem Eq. (14). As an alternative, we apply a generalized variant of the self-consistent Born approximation, wherein  $\mu(\mathbf{x}) = \mu_0 + \sigma^2(\mathbf{x})$ , and  $\sigma(\mathbf{x})$  is a Gaussian distributed variable with zero mean. (In this way, positivity of the elastic constant is ensured.) The actual implementation of the SCBA for this type of disorder turns out to be rather involved and its details will be published elsewhere. Suffice to say that at  $d = 3$ ,  $\alpha_1$  is still positive.  $\alpha_2$ , on the other hand, no longer has definite sign. Applying the high frequency expansion outlined above for the random mass density case we find

$$\alpha_2 = \frac{l^2}{\bar{c}^2 \left\langle m^{\frac{3}{2}} \mu^{-\frac{3}{2}} \right\rangle} \left[ \frac{1}{96} \left\langle \frac{\mu^2 (\nabla m)^2}{m^{\frac{3}{2}} \mu^{\frac{5}{2}}} \right\rangle + \frac{5}{48} \left\langle \frac{m \mu \nabla m \nabla \mu}{m^{\frac{3}{2}} \mu^{\frac{5}{2}}} \right\rangle - \frac{23}{96} \left\langle \frac{m^2 (\nabla \mu)^2}{m^{\frac{3}{2}} \mu^{\frac{5}{2}}} \right\rangle \right]. \quad (15)$$

If  $\alpha_2$  is negative (which happens, for example, in the limiting case of non-random  $m(\mathbf{x})$ )  $f(u)$  no longer has a maximum at  $u \simeq 1$ . Instead,  $I(\omega)$  is an increasing function of  $\omega$  with a broad maximum reached at frequencies  $\omega > \bar{c}/l$ , before dropping off at frequencies higher than  $\omega_D$ . Concluding, we find that, depending on the profile of the disorder, the envelope function  $I(\omega)$  of the random mass/elastic constants problem either contains a low-frequency peak, or a broad high frequency maximum.

So far, we have considered the case of scalar phonons. In a realistic environment, however,  $\psi \rightarrow u_i$  will be a

$d$ -component *vector*. The most general random vector problem would be governed by a formidable rank four random elastic modulus tensor. Assuming, however, a medium consisting of a random accumulation of 'micro-crystallites' each of which possessing intrinsic rotational invariance — this assumption may well be violated, especially in glassy environments, although it seems to work in polycrystalline materials — the effective wave equation reduces to

$$\partial_i [\lambda(\mathbf{x}) \partial_j u_j] + \partial_j [\mu(\mathbf{x}) (\partial_i u_j + \partial_j u_i)] + \omega^2 m(\mathbf{x}) u_i = 0, \quad (16)$$

where  $m(\mathbf{x})$  is a random density of the medium and  $\lambda(\mathbf{x})$ ,  $\mu(\mathbf{x})$  are random Lamé coefficients.

In the limit where only  $\lambda(\mathbf{x})$  is random, Eq. (16) can be mapped into Eq. (5) by substitution of  $\psi = \partial_i u_i (\lambda + 2\mu)$ . In this case, both  $\alpha_1$  and  $\alpha_2$  are positive. This result carries over to the case where, in addition to random  $\lambda(\mathbf{x})$ , also the density  $m(\mathbf{x})$  is made random; For these two types of disorder  $I(\omega)$  has a peak at  $\omega \sim \bar{c}/l$ . If, however, the shear modulus  $\mu$  is also random, Eq. (16) becomes more similar to Eq. (14). Applying the SCBA, it is still possible to show that  $\alpha_1 > 0$  (for  $d > 2$ ). As with our previous discussion of (14), however,  $\alpha_2$  is no longer of definite sign. Specifically,  $I(\omega)$  will contain a broad maximum ( $\alpha_2 < 0$ ) if only the shear modulus  $\mu(\mathbf{x})$  is random.

To conclude, we have shown that, depending on the type of disorder, the normalized DoS  $I(\omega)$  of random wave equations may either contain a peak at phonon wavelengths of the order of the disorder correlation length, or a broad maximum at wavelengths below the correlation length. We believe that this work is not only relevant for the interpretation of data on vibrational modes in random media but also to the analysis of other types of waves, such as electromagnetic waves in random environments.

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